

# Alternative representation of time-dependent Hamiltonians with application to laser-driven systems

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A representation of time-dependent Hamiltonians that describe laser-driven systems is presented. Unlike the well-known time-independent dressed potentials that are functions of the characteristic parameter  $\alpha_0 = \sqrt{I}/\omega^2$ , where  $\omega$  and  $I$  are the laser frequency and intensity, this approach provides a time-averaged potential that depends explicitly on the field parameters; e.g.,  $I$ ,  $\omega$ , and shape of the laser pulse. The modified dressed potential is  $\hbar$  independent and adds a classical time-independent potential barrier to the Kramers-Henneberger dressed potential. We show that this dynamical potential barrier is identical to the Kapitza effective classical potential energy obtained for the motion of a particle in a rapidly oscillating field. As an illustrative numerical example, a simple one-electron effective model Hamiltonian of xenon atom in strong laser field is studied. We show that the zero-order quasienergies obtained by our representation are reasonably accurate and the second order high-frequency perturbation calculations provide quite accurately the lifetime of the photoionized electron for a broad range of laser frequencies.

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## I. INTRODUCTION

The study of a system of an atom or a molecule driven by a strong laser field requires the solution of the time-dependent Schrödinger equation when the Hamiltonian is time-dependent. The numerical exact solution of this equation is usually very complicated and the effort is costly, in particular when the electron correlation is taken into consideration. Such calculations were carried out, for example, for the harmonic generation of helium atom in KrF laser [1] and for double ionization of two-electron atoms [2–7]. Since the solution of the time-independent Schrödinger equation is much simpler than that of the time-dependent one, the question arises whether one can solve the problem of the laser-driven atom/molecule using an approximation where the time-independent Schrödinger equation with an effective potential is solved. Within the framework of the dipole approximation, the equation we need to solve is

$$\hat{H}\Psi = i\hbar \frac{\partial}{\partial t}\Psi, \quad (1)$$

where the Hamiltonian is defined as

$$\hat{H}(q,t) = \hat{H}_{atom/molecule} + e\epsilon_0 q f(t). \quad (2)$$

Here  $f(t)$  gives the time dependence of the electromagnetic radiation. For example, when a cw laser is used, then  $f(t) = \cos \omega t$ . For such periodic driving, it is possible to obtain quasistationary solutions

$$\Psi_j(q,t) = e^{-iE_j t/\hbar} \Phi_j(q,t), \quad (3)$$

where

$$\Phi_j(q,t) = \Phi_j(q,t+T) \quad (4)$$

and  $T = 2\pi/\omega$ . The quasistationary states  $\Phi_j$  are solutions of the Floquet Hamiltonian

$$\hat{H}_f = -i\hbar \frac{\partial}{\partial t} + \hat{H}(q,t), \quad (5)$$

$$\hat{H}_f \Phi_j(q,t) = E_j \Phi_j(q,t), \quad (6)$$

where  $E_j$  are the quasienergies (QE) of the system. The quasienergies are defined mod  $\hbar\omega$ , meaning that  $E_j + n\hbar\omega$  (where  $n$  is any integer), is also an eigenvalue of the Floquet Hamiltonian. Note that this approach has been generalized by the  $(t, t')$  method for the most general case where the field is not necessarily time periodic [8]. That is, even when the Hamiltonian is not time periodic, e.g., when short laser pulses are used, then the time-dependent solution of the Schrödinger equation is given by

$$\Psi(t) = e^{-i\hat{H}_f(q,t')t/\hbar} \Psi(0), \quad (7)$$

when  $\hat{H}_f$  is a Floquet-type operator [as defined in Eq. (5)] and  $0 \leq t \leq T$ .  $T$  in this case is any time that is longer than the duration of the laser pulse;  $t'$  serves as an additional coordinate that makes the evolution operator time independent. This approach enables the use of the Floquet theory for cases when the Hamiltonian is not time periodic. The spectrum of  $\hat{H}_f$  is a continuous one and unbound from below. Therefore it does not support bound states. The QE are associated with both continuum states and resonance states [9]. The resonances are metastable states for which the electrons are trapped by the localized potential. In Hermitian quantum mechanics the resonances are associated with a localized wave packet. When outgoing boundary conditions are imposed on the solution of the Schrödinger equation (known as the Siegert boundary conditions) [10], the resonances are associated with complex eigenvalues  $E_j = \epsilon_j - (i/2)\Gamma_j$ , where  $\epsilon_j$  is the

energy position and  $\Gamma_j$  is the width of the resonance state. In order to avoid the need to impose outgoing boundary conditions and in order to bring the resonances into the generalized Hilbert space (as bound states in Hermitian QM), the complex scaling method has been introduced [11,12]. Upon complex scaling,  $q \rightarrow q \exp(i\theta)$ , the resonance wave function becomes square integrable. However, one should be aware of the generalization of the definition of inner product when the Hamiltonian is non-Hermitian [13,12]. The resonances are the complex eigenvalues that are not effected by the change in  $\theta$  (provided  $\theta$  exceeds a critical value).

## II. TIME-INDEPENDENT HIGH-FREQUENCY PERTURBATION THEORY FOR DRIVEN SYSTEMS IN STRONG LASER FIELDS

Using the  $(t, t')$  method, time is defined as an additional coordinate [8], and therefore time independent perturbation theory is applicable [8,14]. Since the radius of convergence is zero even when weak ac/dc fields are applied we deal with asymptotic perturbational series. For strong laser field, the question what is  $\hat{H}_0$  is a crucial one. When the laser frequency  $\omega$  is smaller than the field-free electron motion frequency  $\Omega_e^{FF}$  (defined as the first excitation energy divided by the Planck constant), time can be treated as an adiabatic parameter. Therefore, when  $\omega/\Omega_e^{FF} \ll 1$ , in the first step of the adiabatic calculations one should calculate the eigenfunctions of  $\hat{H}(q,t) = \hat{H}_0^{FF} + e\epsilon_0 q f(t)$ , where  $\hat{H}_0^{FF}$  is the field-free Hamiltonian  $\hat{H}_0^{FF} = \hat{p}^2/2\mu + V(q)$ ,  $\epsilon_0$  is the maximum field amplitude, and  $f(t)$  is the time-dependent part of the electric field. In this case the natural choice for the zeroth-order Hamiltonian is the field-free one,  $\hat{H}_0^{FF}$ . However, when  $\omega/\Omega_e^{FF} \gg 1$ , the electrons move in the time-averaged effective potential. When  $\hat{H}_0$  is the field-free Hamiltonian, then, the time-averaged potential is zero for periodically driven systems. In this case it is preferable to apply perturbation theory after carrying out the transformation to the acceleration frame [known as Kramers-Henneberger (KH) representation] [15]. In this representation the origin of the coordinates oscillates with the field. The transformed Hamiltonian has the form

$$\hat{H}_f^{KH} = -i\hbar \frac{\partial}{\partial t} + \frac{\hat{p}^2}{2\mu} + V\left(q + \frac{e\epsilon_0}{\mu} \int \int dt f(t)\right). \quad (8)$$

For cw lasers,  $f(t) = \cos \omega t$ ,  $V(q - \alpha_0 \cos \omega t)$ , and  $\alpha_0 = e\epsilon_0/\mu\omega^2$ . In order to simplify the formulation, we denote  $V(q - \alpha_0 \cos \omega t)$  as  $V^{KH}(q,t)$ :

$$V^{KH}(q,t) \equiv V(q - \alpha_0 \cos \omega t). \quad (9)$$

Under this transformation the zero-order Hamiltonian  $H_0 = \hat{T} + V_0^{KH}$  depends only on  $\alpha_0$  and not explicitly on the laser frequency and the maximal field amplitude. The zero-order effective potential is defined as

$$V_0^{KH} = \frac{1}{T} \int_0^T V^{KH}(q,t) dt. \quad (10)$$

It is clear that the zero-order perturbation theory is applicable only when the laser frequency is much larger than the frequency of electronic oscillations inside the effective potential well,  $\Omega_e$  [18]. From Eq. (10) one can see that as  $\alpha_0$  gets larger values, the zeroth order effective potential  $V_0$  becomes shallower. Therefore, the value of  $\Omega_e$  is reduced as the parameter  $\alpha_0 = e\epsilon_0/\mu\omega^2$  is increased. The condition  $\omega \gg \Omega_e$  implies that this perturbation theory scheme holds only for very strong laser fields, and when the laser frequency is sufficiently high. Therefore, for sufficiently strong laser fields the quasienergies can be calculated by the diagonalization of a time-independent Hamiltonian  $\hat{H}_0 = \hat{T} + V_0^{KH}$ , where  $V_0^{KH}$  is defined in Eq. (10). This result enables the use of standard computational chemistry programs for the calculation of the zero-order quasienergy values of atoms in high-intensity laser fields [16]. In their calculations they had considered the multipole expansion of the exact Coulomb electron-nucleus interaction, as given in Ref. [17]. The same approach can be taken using the expression for the time-independent Hamiltonian presented in the following Section. The time-independent expression for the time-independent Hamiltonian enables the calculations of the dependence of the quasienergy spectrum on the laser frequency and independently, on the intensity of the laser. Note by passing that the quasienergy spectrum is a measurable quantity. Such measurements can provide information on the nonlinearity of the photoinduced dynamics of the studied system and its dependence on the laser field parameters.

Let us solve the Floquet eigenvalue problem  $\hat{H}_f^{KH} \Phi_j = E_j \Phi_j$  (where  $\Phi_j$  are time periodic functions) using perturbation theory. The  $n$ th order QE are calculated using the time-independent perturbation theory when time is treated as an additional coordinate. Taking this approach, the higher order perturbational terms are calculated when the time-dependent potential is expanded in Fourier basis. Namely,

$$V_{pert} = V^{KH}(q,t) - V_0^{KH} = \sum_{m \neq 0} V_m e^{im\omega t}, \quad (11)$$

where

$$V_m(q) = \frac{1}{T} \int_0^T e^{-im\omega t} V^{KH}(q,t) dt. \quad (12)$$

Note that the zeroth Fourier component is subtracted from the perturbation since it is included in the zero-order Hamiltonian. The zero-order quasistationary solutions are given by

$$\hat{H}_0 \Phi_j^{(0)} = E_j^{(0)} \Phi_j^{(0)}. \quad (13)$$

The first-order corrections to the quasienergies vanish because of the integration over the "time"  $t'$  coordinate. The second-order corrections to the quasienergies are given by

$$E_j^{(2)} = \sum_{m \neq 0} \sum_k \frac{(\Phi_k^{(0)} | V_m | \Phi_j^{(0)})^2}{E_j^{(0)} - (E_k^{(0)} + \hbar \omega m)}. \quad (14)$$

The expressions given above are valid for many-electron systems where the electron correlation is taken into account.

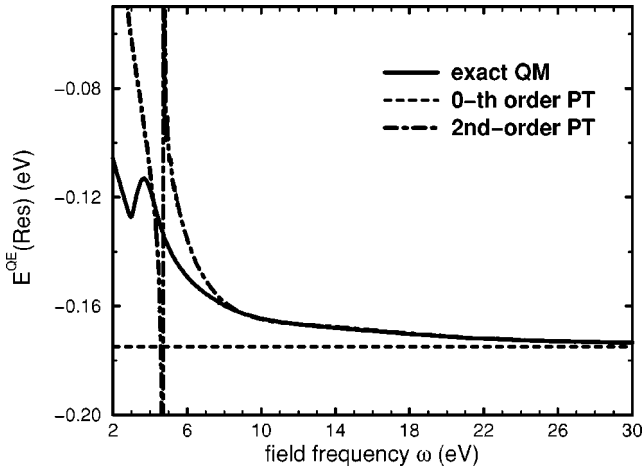


FIG. 1. Exact, zero-order, and second-order quasienergies obtained by the use of the KH representation of the ground-state quasienergies of xenon as function of the laser frequency. The maximum field amplitude is varied to keep  $\alpha_0 = 6$  a.u.

The round bra-ket  $(\dots)$  stands for the  $c$  product rather than the regular scalar product when complex scaling is used [12]. If the eigenfunctions of the unscaled  $\hat{H}_0$  are real, then  $(f|g) = (f^*|g)$ . Note that such high-order corrections to this high-frequency Floquet theory have been calculated before [19]. We have chosen, for the sake of simplicity, as an illustrative model Hamiltonian a one-dimensional effective potential for the ionization of Xe [20].

$$V(q) = -D_0 \exp(-\gamma q^2), \quad (15)$$

where  $D_0 = 0.63$  a.u. and  $\gamma = 0.1424$  a.u. A comparison between the ground-state quasienergy obtained by this perturbation theory and the exact QM calculations performed by the  $(t, t')$  method show that only in the very high frequency regime, the zero-order result converges to the exact one (Fig. 1). We would like to emphasize that, using this approach the zero-order quasienergies are not a function of the laser frequency  $\omega$  but of  $\alpha_0 = e\epsilon_0/\mu\omega^2$ . The results of the second order perturbation calculations presented in Fig. 1 show that the correct quasienergies are obtained by second-order perturbation theory when the laser frequency  $\omega$ , is larger than the ionization energy  $EI$ . The transition from high-frequency ionization mechanism (i.e., when single-photon ionization is dominant) to the low-frequency ionization mechanism (in which ionization is due to multiphoton absorption) takes place when  $\hbar\omega \sim IE$ .

Widely used software packages for electron-structure calculations (such as Gaussian) can be used for calculating ground electronic states only. Consequently, they are applicable within the framework of the zero-order perturbation theory described above. Therefore, one may wonder how perturbation theory should be used in order to include explicitly the dependence on the laser frequency in the zero-order perturbational calculations. This subject will be discussed in the following section.

### III. PERTURBATION THEORY USING AN ALTERNATIVE REPRESENTATION OF TIME-DEPENDENT HAMILTONIANS OF DRIVEN SYSTEMS IN STRONG LASER FIELDS

In the approach we present here, the Hamiltonian given in Eq. (8) is transformed once again by the following transformation:

$$\Phi_j^{alt}(q, t) = \exp\left[(i/\hbar) \int^t [V^{KH}(q, t') - V_0^{KH}] dt'\right] \Phi_j^{KH}(q, t). \quad (16)$$

The motivation for such a transformation is to obtain a time-averaged potential with a better dependence on the field parameters. As a result of this transformation, the Hamiltonian is given by

$$\hat{H}_f^{alt}(q, t) = \hat{H}_0^{KH} + V^{alt}(q, t) - i\hbar \frac{\partial}{\partial t}, \quad (17)$$

where

$$V^{alt}(q, t) = -\frac{1}{2\mu\omega^2} \left( \sum_{n \neq 0} \frac{f_n}{n} e^{in\omega t} \right)^2 - \frac{\hbar}{2\mu\omega} \sum_{n \neq 0} \frac{1}{n} \frac{\partial f_n}{\partial q} e^{in\omega t} - \frac{\hbar}{\mu\omega} \sum_{n \neq 0} \frac{f_n}{n} e^{in\omega t} \frac{\partial}{\partial q}. \quad (18)$$

Here  $f_n(q)$  is the  $n$ th Fourier component of the force

$$f_n(q) = -\frac{\partial V_n(q)}{\partial q}. \quad (19)$$

These Fourier components of the force play important role in the generation of high-order harmonics in the high-frequency regime [21]. This potential can also be represented in its Hermitian form

$$V^{alt}(q, t) = \frac{1}{2\mu} [F^2(q, t) + \hat{p}F(q, t) + F(q, t)\hat{p}], \quad (20)$$

where

$$F(q, t) = - \int \frac{\partial V_{KH}(q, t)}{\partial q} dt = \sum_{n \neq 0} \frac{f_n(q)}{n\omega} e^{in\omega t}. \quad (21)$$

This new Hamiltonian seems rather complicated. When we apply perturbation theory to it, we obtain an expression for a zero order potential, which has a simple form:

$$V_0^{alt}(q) = V_0^{KH} + \frac{1}{T} \int_0^T V^{alt}(q, t) dt = V_0^{KH} + \frac{1}{2\mu\omega^2} \sum_{n \neq 0} \frac{f_n(q)f_{-n}(q)}{n^2}. \quad (22)$$

It is important to emphasize the dependence of the time-independent dressed potential given in Eq. (22) on the frequency of the laser (see Fig. 2). Clearly, the contribution of

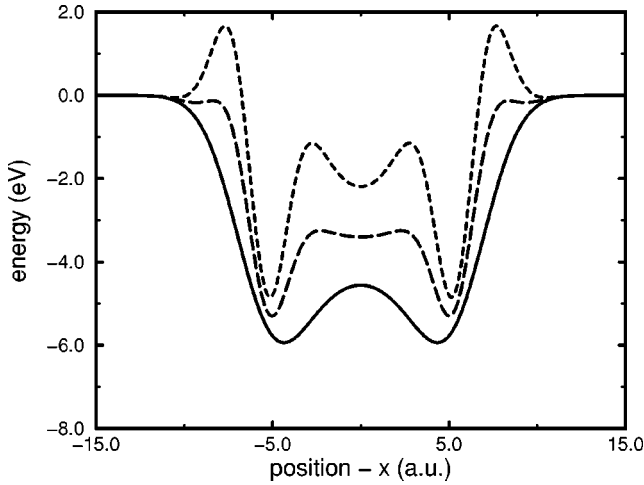


FIG. 2. Effective time-averaged potentials for the xenon atom in a strong laser field when  $\alpha_0 = e\epsilon_0/\mu\omega^2 = 6$ . Solid line, KH dressed potential; long-dashed line, dressed potential when  $\omega = 5$  eV; dashed line, dressed potential when  $\omega = 3.5$  eV.

this term to the perturbational expansion is increased as the frequency  $\omega$  is decreased. The results of the zero-order quasienergies calculated for the model Hamiltonian, which describes the photoionization of a model xenon atom, are presented in Fig. 3. These results show that the accuracy of the calculations of the quasienergy spectrum by zeroth order perturbation theory is drastically improved when the perturbational approach is taken rather than the “conventional” one (i.e., using the Kramers-Henneberger representation). The transformation presented in this paper adds to the “conventional” time-independent dressed potential a term that is defined in Eq. (22). This term does not depend on the Planck constant. For cw lasers, this potential term is definitely positive and can be regarded as a classical (i.e.,  $\hbar$ -independent) time-independent potential barrier. In Fig. 2, we show the difference between the KH dressed potential and the new

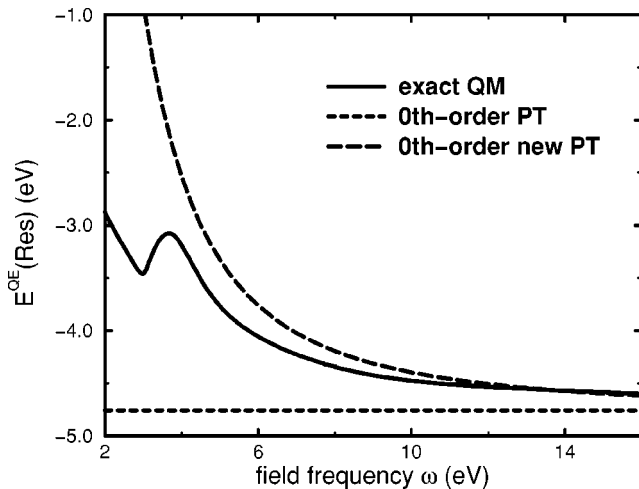


FIG. 3. Comparison between the exact and zero-order quasienergies obtained by the use of the KH representation and our representation. The maximum field amplitude is varied to keep  $\alpha_0 = 6$  a.u.

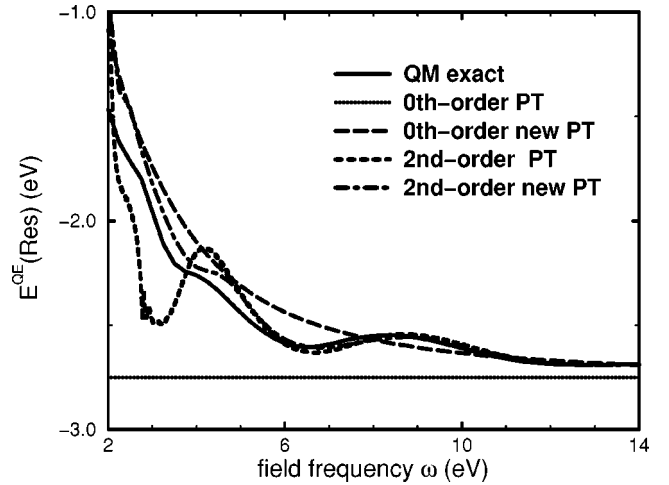


FIG. 4. Comparison between the exact, zero-order, and second-order quasienergies obtained by the use of the KH representation and our representation. The maximum field amplitude is varied to keep  $\alpha_0 = 12$  a.u.

dressed potential as  $\omega$  is varied at fixed  $\alpha_0$ . It is easy to see that this potential barrier is exactly equal to the time average of  $[F(q,t) - f_0(q)]^2 / (2\mu\omega^2)$ , where  $f_0(q)$  is the force of the dressed potential,  $f_0(q) = -dV_0^{KH}(q)/dq$ . This is the Kapitza effective classical potential energy obtained for the motion of a particle in a rapidly oscillating field [22]. Our method can be extended by replacing the time-averaged potential  $V_0 \equiv V^{KH}$  that appears in the transformation given in Eq. (16) by a different time-independent potential. The chosen  $V_0$  potential used in the transformation should provide the best agreement between the eigenvalues of the time-independent problem and the exact QE. The criteria for choosing  $V_0$  is an open question and outside the scope of this paper. In this manner, it might be possible to achieve different functional dependences of the zero-order Hamiltonian on  $\omega$ . This might be the direction one should take in order to extend the applicability of the zero-order perturbation theory presented here to a broad range of laser frequencies.

In Fig. 4 we represent the quasienergies as a function of the laser frequency (while the parameter  $\alpha_0$  is held fixed) as obtained from our second-order perturbation theory calculations. As one can see, the quasienergies up to the second order, which are obtained by using our approach, are more accurate than those obtained by using perturbation theory in KH frame. However, the accuracy of quasienergies calculations is not improved drastically by the use of the second-order perturbation theory as in the zero-order calculations. This is due to the fact that in the Kramers-Henneberger representation, unlike the zero-order quasienergies, the second-order quasienergies depend explicitly on the laser frequency. In our calculations, the parameter  $\alpha_0$  was held fixed as the laser frequency  $\omega$  was varied. It implies that the laser intensity (proportional to the square of the maximum field amplitude  $\epsilon_0$ ) has been increased with the increase of  $\omega$ .

As been stated before, the QE states have a finite lifetime. The inverse of the lifetime of a resonance quasienergy state is associated with its width (which is represented by the imaginary part of the quasienergy). In our studied case, the

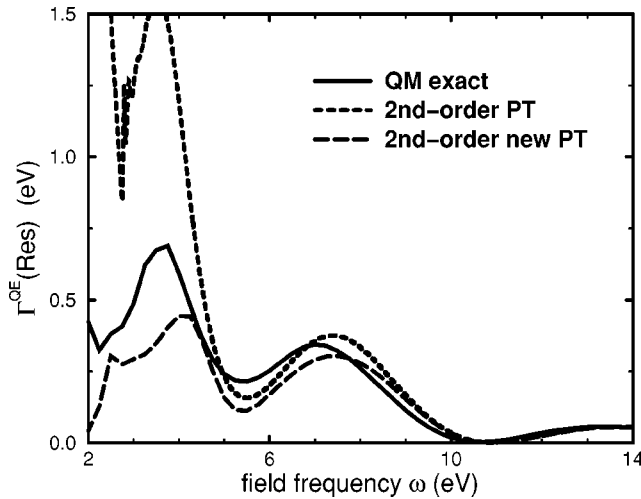


FIG. 5. Comparison between the exact and second-order resonance width (inverse lifetime) obtained by the use of the KH representation and our representation. The maximum field amplitude is varied to keep  $\alpha_0 = 12$  a.u.

ground resonance width cannot be calculated within the framework of the zero-order perturbation theory, since the ground state of the effective time-independent dressed potential is a bound state. The ground-state resonance width, however, can be obtained by using the second-order perturbation theory as shown in Fig. 5. As one can see from the results presented in Fig. 5, when the laser frequency is below 10 eV then the perturbation approach presented here gives more accurate resonance widths than those obtained by the perturbation theory in KH frame. It also provides the qualitative overall frequency behavior of the width from 2.5 eV (KrF laser) and above.

#### IV. CONCLUSIONS

Zero-order perturbation theory in the KH (Kramers-Henneberger) frame provides a time-independent dressed potential that varies with the characteristic parameter  $\alpha_0$ . When the laser frequency is sufficiently large, the use of the KH dressed potential enables one to solve the time-independent Schrödinger equation rather than the time-dependent equation. This time-independent approach is in particular attractive when the electronic correlation effects are considered and the electron-structure calculations are carried out with the help of commercial program packages [16]. However, the

quasienergies that are obtained using this approach are function of one variable only. It is impossible, for example, to use the KH-dressed potentials for studying the effect of the variation of the laser frequency and independently of the laser intensity on the quasienergy spectrum of the studied problem. Our approach based on an alternate transformation of the time-dependent Schrödinger equation provides a modified dressed potential that is applicable not only for high frequencies but also for a broad range of frequencies of lasers commonly used today in the study of nonlinear optical phenomena (such as high-order harmonic generation). The main advantage of this approach is that it provides a zero-order time-independent dressed potential that explicitly depends on all laser parameters. This is particularly important in the study of the effect of field parameters (e.g., duration of the laser pulse, its frequency, and intensity) on the quasienergy spectrum (or other properties) of the laser-driven systems. However, one still has to prove that the perturbative expansion obtained by the transformation in Eq. (16) is convergent at low laser frequencies.

We have shown that for periodically driven systems, a modified dressed potential is obtained resulting from the transformation presented here. The new dressed potential presented here differs from the KH potential by a potential barrier. This barrier that is  $\hbar$  independent is shown to be identical to the Kapitza effective classical potential energy obtained for the motion of a particle in rapidly oscillating field. It is expected that this time-independent classical potential barrier will induce temporarily trapping of particles (so-called narrow resonances). This kind of temporary trapping of particles is different from the light-induced states that are discussed extensively in the literature [23,24]. The light-induced states are bound states in the dressed potential picture, whereas the temporarily trapped states due to the existence of Kapitza potential barrier have a finite lifetime even in the dressed picture. The conditions that lead to the appearance of such states is under current study.

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